Search for α decay of naturally occurring Hf-nuclides using a Cs_2HfCl_6 scintillator

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on behalf of the collaboration.
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Potentially α decay of naturally occurring Hf-nuclides

Some potential transitions of Hf isotopes and related information. Only naturally occurring isotopes (with natural abundance δ) and with Q > 0 between g.s. transitions or between g.s. and lowest bound level transitions (with spin/parity J^π) are listed. E is the kinetic energy of the alpha particle. N is the number of nuclei in the CHC crystal used in this work. Experimental measurements (when available) and theoretical prediction of the half-live are reported in the last four columns.

Nuclide Transition	J^π of Parent → Daughter Nuclei and its level (keV) [10, 11]	δ (%) [2]	Q_α (keV) [12]	E_α (keV)	N	$T_{1/2}$ (y) Experimental	$T_{1/2}$ (y) Theoretical [15]	Theoretical [16]	Theoretical [9]
$^{174}Hf \rightarrow ^{170}Yb$	$0^+ \rightarrow 0^+$, g.s. $0^+ \rightarrow 2^+$, 84.2	0.16(12)	2494.5(2.3)	2437.6(2.2)	1.0×10^{19}	$2.0(4) \times 10^{15}$ [6, 13] $\geqslant 3.3 \cdot 10^{18}$ [14]	$3.5 \cdot 10^{16}$ $1.3 \cdot 10^{18}$	7.4×10^{16} 3.0×10^{18}	3.5×10^{16} 6.6×10^{17}
$^{176}Hf \rightarrow ^{172}Yb$	$0^+ \rightarrow 0^+$, g.s. $0^+ \rightarrow 2^+$, 78.7	5.26(70)	2254.2(1.5)	2203.3(1.5)	3.3×10^{20}	– $\geqslant 3.0 \times 10^{17}$ [14]	2.5×10^{20} 1.3×10^{22}	6.6×10^{20} 3.5×10^{22}	2.0×10^{20} 4.9×10^{21}
$^{177}Hf \rightarrow ^{173}Yb$	$7/2^- \rightarrow 5/2^-$, g.s. $7/2^- \rightarrow 7/2^-$, 78.6	18.60(16)	2245.7(1.4)	2195.3(1.4)	1.2×10^{21}	– $\geqslant 1.3 \times 10^{18}$ [14]	4.5×10^{20} 9.1×10^{21}	5.2×10^{22} 1.2×10^{24}	4.4×10^{22} 3.6×10^{23}
$^{178}Hf \rightarrow ^{174}Yb$	$0^+ \rightarrow 0^+$, g.s. $0^+ \rightarrow 2^+$, 76.5	27.28(28)	2084.4(1.4)	2037.9(1.4)	1.7×10^{21}	– $\geqslant 2.0 \times 10^{17}$ [14]	3.4×10^{23} 2.4×10^{25}	1.1×10^{24} 8.1×10^{25}	2.2×10^{23} 7.1×10^{24}
$^{179}Hf \rightarrow ^{175}Yb$	$9/2^+ \rightarrow 7/2^+$, g.s. $9/2^+ \rightarrow 9/2^+$, 104.5	13.62(11)	1807.7(1.4)	1767.6(1.4)	8.6×10^{20}	$\geqslant 2.2 \times 10^{18}$ [14] $\geqslant 2.2 \times 10^{18}$ [14]	4.5×10^{29} 2.0×10^{32}	4.0×10^{32} 2.5×10^{35}	4.7×10^{31} 2.2×10^{34}
$^{180}Hf \rightarrow ^{176}Yb$	$0^+ \rightarrow 0^+$, g.s. $0^+ \rightarrow 2^+$, 82.1	35.08(33)	1287.1(1.4)	1258.7(1.4)	2.2×10^{21}	– $\geqslant 1.0 \times 10^{18}$ [14]	6.4×10^{45} 4.0×10^{49}	5.7×10^{46} 4.1×10^{50}	9.2×10^{44} 2.1×10^{48}

T.P. Kohman, Phys. Rev. 121, 1758 (1961);

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The experiment

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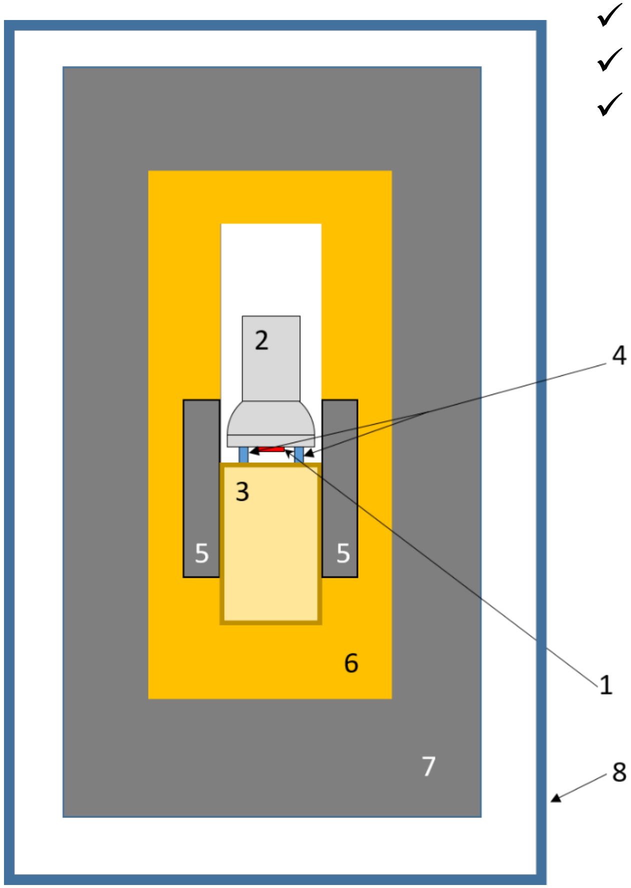
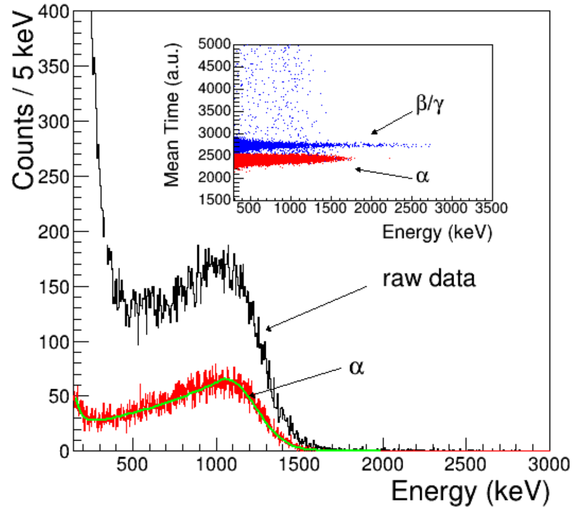
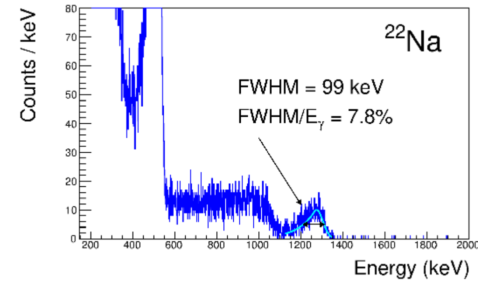
- ✓ Cs_2HfCl_6 crystal (CHC) 6,90(1) g
- ✓ CHC crystal coupled low-radioactivity PMT (Hamamatsu R6233MOD)
- ✓ placed above the end-cap of the ultra-low background HP-Ge
- ✓ CAEN DT5720B digitizer 250 MSamples/s;
- ✓ 2848 h data taking

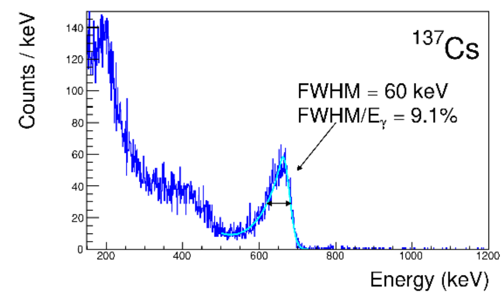


Energy Calibration

Preliminary quenching factor (QF) estimation







Schematic cross-sectional view of the experimental set-up (not in scale). There are shown the CHC crystal scintillator (1) coupled with a 3 inches PMT (2), the HP-Ge detector (3), which is separated by a cylindrical Teflon ring (4). They are completely surrounded by a passive shield made by archaeological Roman lead (5), high purity copper (6), low radioactive lead (7). The whole set-up (with the exception of the cold finger for the HP-Ge detector) is enclosed in a Plexiglas box (8) continuously flushed with HP-N_2 gas.

Low background measurements of the CHC crystal

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Isotopic composition of ^{nat}Hf measured in a sample of the CHC crystal by ICP-MS

Isotope	Abundance (%)
^{174}Hf	0.156(6)
^{176}Hf	5.18(5)
^{177}Hf	18.5(1)
^{178}Hf	27.2(1)
^{179}Hf	13.9(1)
^{180}Hf	35.2(2)

Concentrations of trace contaminants in the CHC crystal as measured by ICP-MS analysis. The limits are at 68% C.L.

Nuclide	Concentration (ppb)
^{144}Nd	<2.4
^{147}Sm	0.6(1)
^{148}Sm	0.4(1)
^{151}Eu	19(7)
^{152}Gd	<0.02
^{180}W	<0.4
^{184}Os	<0.003
^{186}Os	<0.25
^{190}Pt	<0.02
^{209}Bi	[illegible]

Nuclide	Q_α (keV) [10]	$T_{1/2}$ (y) [10]	Isotopic Abundance (%) [2]	E_α (keV)	Expected Counts
^{144}Nd	1906.4(17)	$2.29(16)\times10^{15}$	23.798(19)	1854.8(17)	<0.007
^{147}Sm	2311.2(10)	$1.060(11)\times10^{11}$	15.00(14)	2249.9(10)	36(6)
^{148}Sm	1986.9(10)	$7(3)\times10^{15}$	11.25(9)	1934.6(10)	$3.6(1)\times10^{-4}$
^{152}Gd	2204.4(10) [11]	$1.08(8)\times10^{14}$	0.20(3)	2147.8(10)	$<1\times10^{-3}$
^{186}Os	2820.4(13)	$2.0(11)\times10^{15}$	1.59(64)	2761.0(13)	$<6\times10^{-4}$
^{190}Pt	3252.6(6)	$6.5(3)\times10^{11}$	0.012(2)	3185.5(6)	< 0.1
^{209}Bi	3137.3(8)	$2.01(8)\times10^{19}$	100	3078.4(8)	$<4\times10^{-7}$

Low background measurements of the CHC crystal

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Isotopic compositio... measured in a samp... CHC crystal by ICP-M...

Isotope	Abunda...
^{174}Hf	
^{176}Hf	
^{177}Hf	
^{178}Hf	
^{179}Hf	
^{180}Hf	

...nts in the CHC ...lysis. The limits

...n (ppb)
<2.4
0.6(1)
0.4(1)
19(7)
<0.02
<0.4
<0.003
<0.25
<0.02

Radioactive contaminations of the CHC crystal measured with the ultra-low background HP-Ge spectrometer GeCris of the STELLA facility at LNGS.

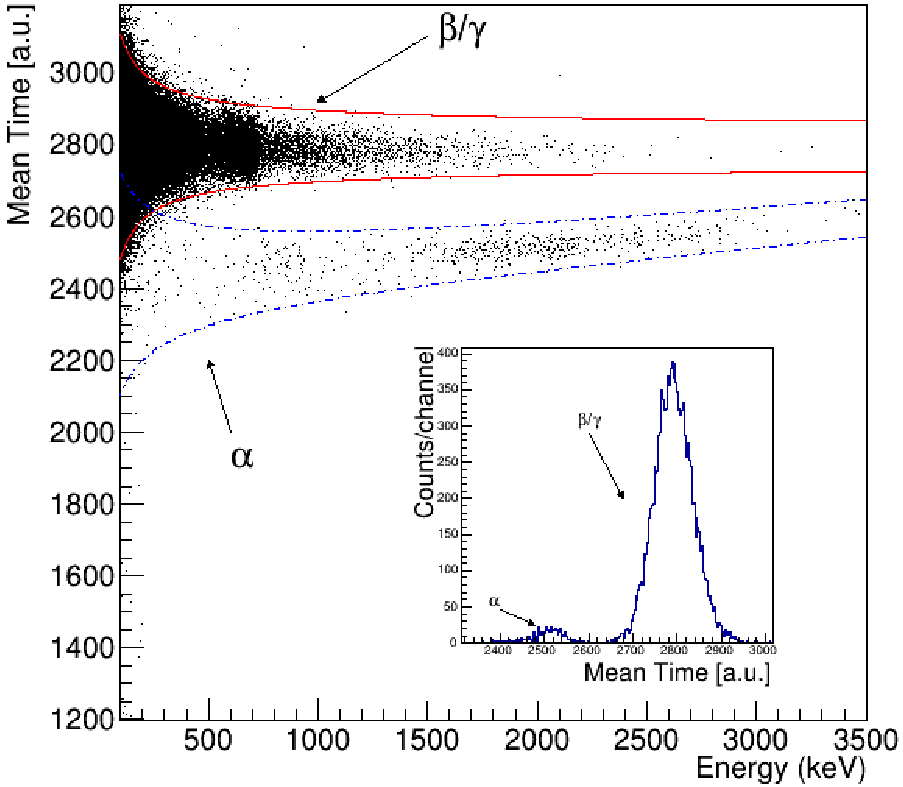
Chain	Nuclide	Activity (mBq/kg)
	^{40}K	$0.4(1)\times10^3$
	^{44}Ti	10(4)
	^{60}Co	<25
	^{137}Cs	$0.74(8)\times10^3$
	^{132}Cs	<15
	^{134}Cs	79(8)
	^{181}Hf	<11
	^{190}Pt	<20
	^{202}Pb	<9.1
^{232}Th	^{228}Ra	<12
	^{228}Th	<3.6
^{238}U	^{226}Ra	<23
	^{234}Th	<0.80
	^{234m}Pa	<0.48
^{235}U	^{235}U	<14

Nuclide					...ected ...unts
^{144}Nd					...007
^{147}Sm					...(6)
^{148}Sm					$\times 10^{-4}$
^{152}Gd					$\times 10^{-3}$
^{186}Os					$\times 10^{-4}$
^{190}Pt					...0.1
^{209}Bi	3137.3(8)	$2.01(8)\times10^{19}$	100	3078.4(8)	$< 4\times10^{-7}$

Data analysis

Time-amplitude analysis of ^{228}Th sub-chain and the derived Q.F.

Pulse Shape Discrimination (PSD) based on the pulse mean-time



The time-amplitude analysis was used to select the events of the following decay sub-chain of the **^{232}Th family**:

^{224}Ra (Q = 5789 keV; $T_{1/2}$ = 3.66 d) → **^{220}Rn** (Q = 6405 keV; $T_{1/2}$ = 55.6 s) → **^{216}Po** (Q = 6906 keV; $T_{1/2}$ = 0.145 s) → **^{212}Pb**.

An average activity of ^{228}Th in the CHC crystal scintillator has been estimated:
100(50) μBq/kg

The energies of the peaks of **^{224}Ra**, **^{220}Rn** and **^{216}Po**, selected by the described time-amplitude analysis, are **2260(200) keV**, **2540(200) keV**, **2780(240) keV** (γ scale), respectively.

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Data analysis

Q.F.

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was used to
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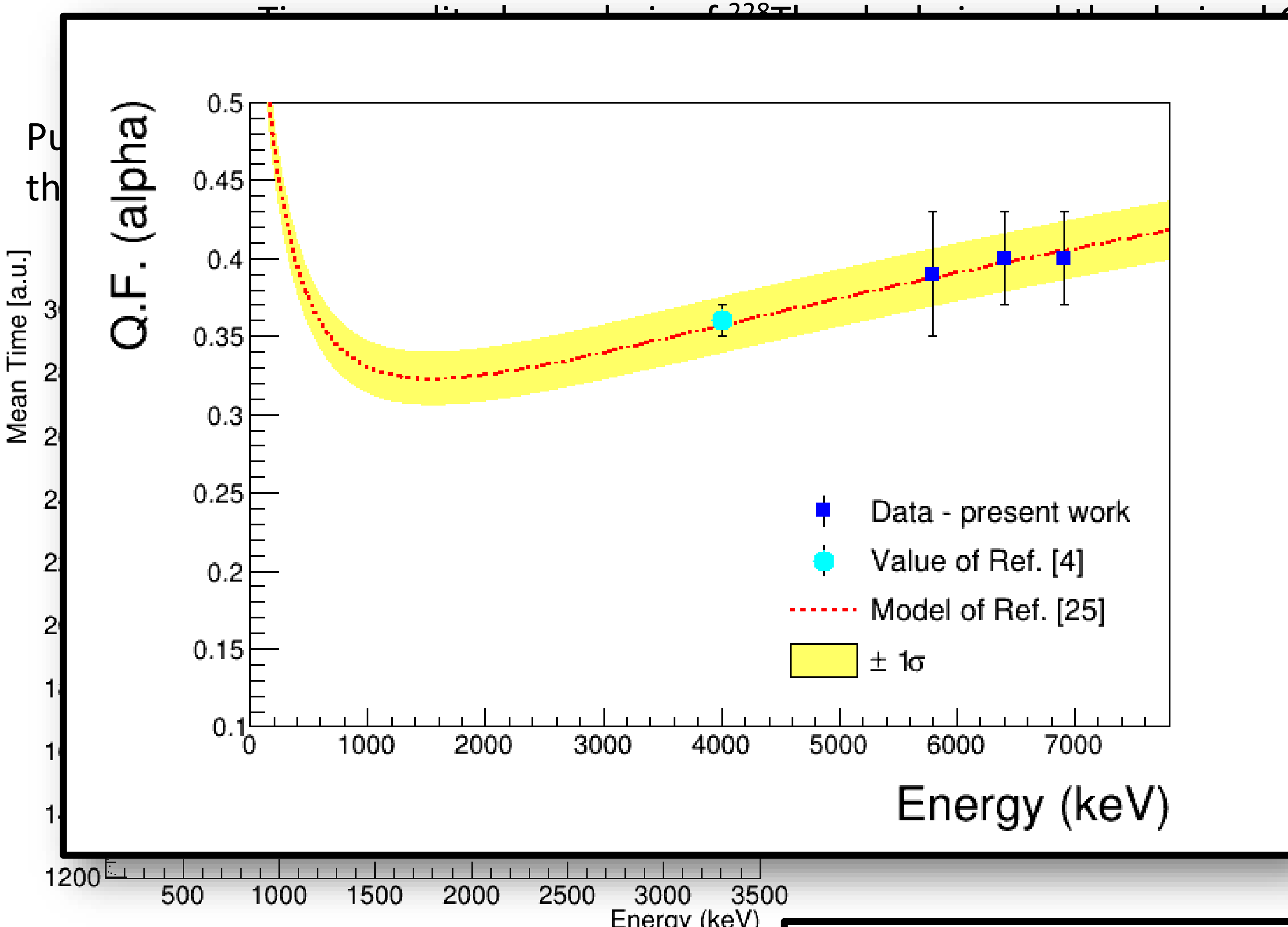
6 d) → ^{220}**Rn** (Q =
Po (Q = 6906 keV;

the CHC crystal
d:



The energies of the peaks of ^{224}**Ra**, ^{220}**Rn** and ^{216}**Po**, selected by the described time-amplitude analysis, are **2260(200) keV**, **2540(200) keV**, **2780(240) keV** (γ scale), respectively.

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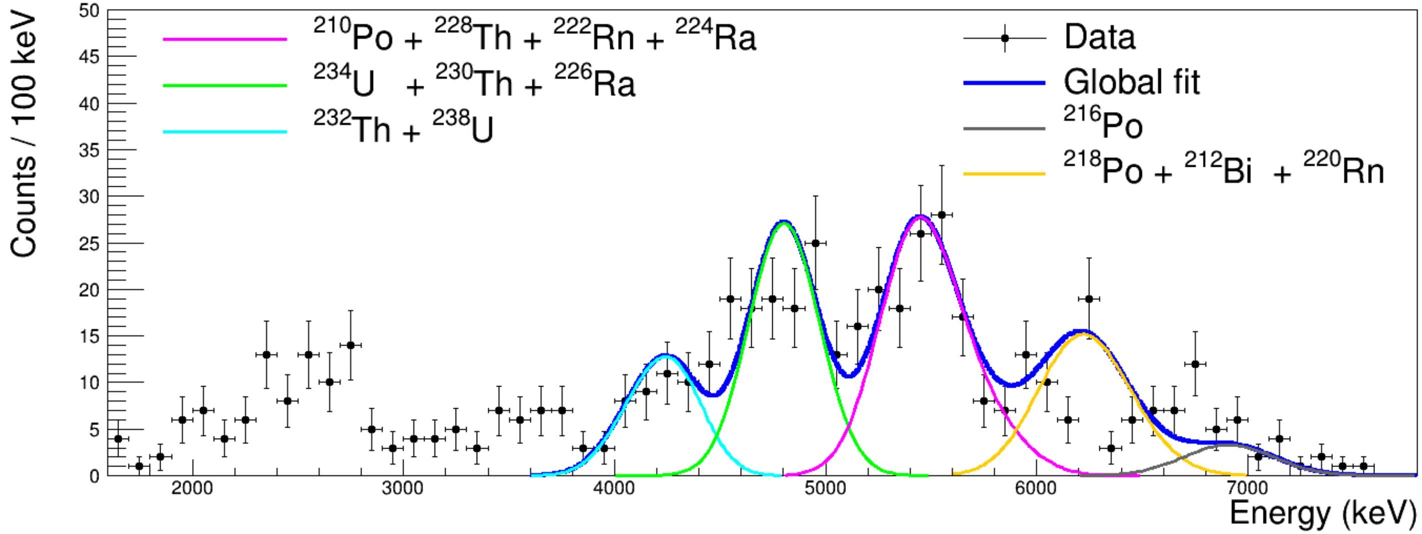
Results on the decay of naturally occurring Hf isotopes



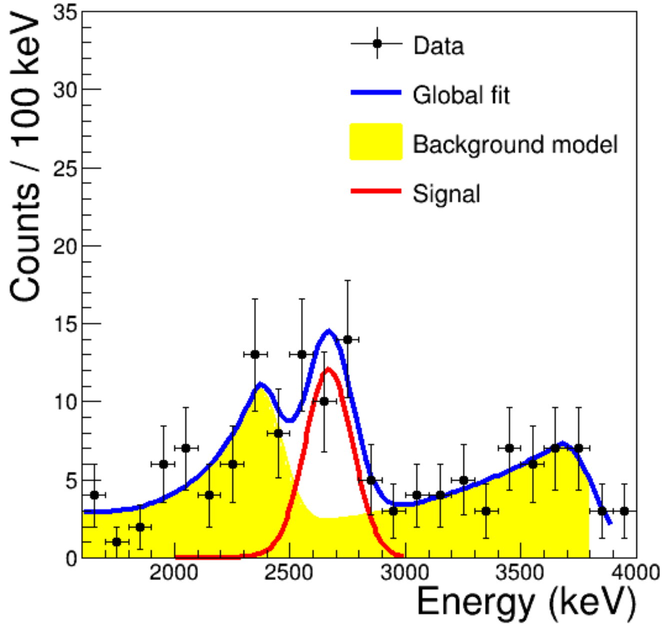
Chain	Sub-Chain	Activity (mBq/kg)
^{232}Th	^{232}Th	0.2(1)
	^{228}Th	0.2(1)
^{238}U	^{238}U	0.6(1)
	^{234}U + ^{230}Th	1.4(2)
	^{226}Ra	0.2(1)
	^{210}Po	1.4(2)

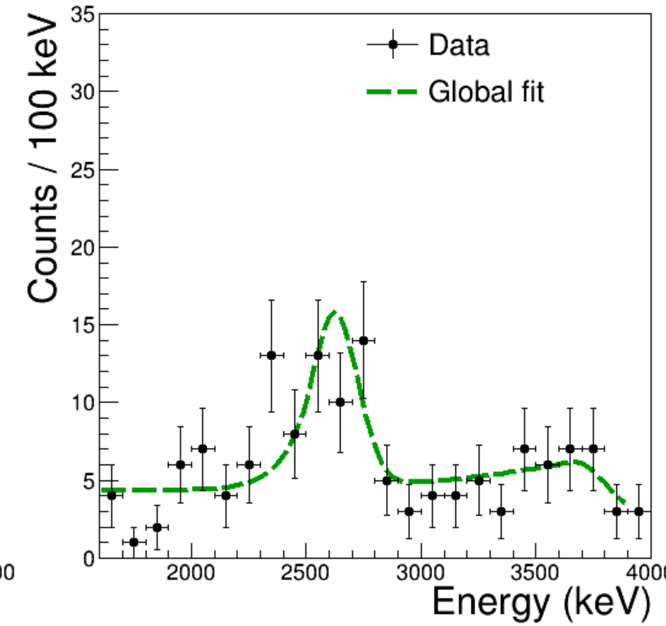
When adopting the claimed half-life 2.0(4) 10^{15}y for the ^{174}Hf decay, the expected number of events - within 2848 h of data taking with the used CHC crystal - is about **1100** counts. Thus, considering that the measured events are **553(23)** in total, even ascribing all of them to ^{174}Hf decay (despite the analysis reported above), one can safely rule out the result of 2.0(4) 10^{15}y; in fact, even in such an unlike hypothesis, the $T_{1/2}$ value derived from the present experimental data would be 4,01(17) 10^{15} y, i.e. is about **4.5 σ far from the value 2.0(4) 10^{15}y**. Thus, the **$T_{1/2}$ value 2.0(4) 10^{15}y is safely rejected**. Let us now perform a more refined determination of the $T_{1/2}$ value of the ^{174}Hf decay supported by our data.

Results on the decay of naturally occurring Hf isotopes

χ^2/n.d.f.=0.87
P-value = 38.7%

Running-test,
tail probabilities:
Upper 94%
Lower 12%





χ^2 probability:
1.7%

Nuclide Transition	Parent, Daughter Nuclei and its Energy Level (keV)	$T_{1/2}$ (y) Experimental: present work	Experimental: previous works [14]	Theoretical [15]	Theoretical [16]	Theoretical [9]
^{174}Hf → ^{170}Yb	$0^+ \to 0^+$, g.s.	$7.0 \pm 1.2 \times 10^{16}$	$2.0 \pm 0.4 \times 10^{15}$ [6, 13]	$3.5 \cdot 10^{16}$	7.4×10^{16}	3.5×10^{16}
	$0^+ \to 2^+$, 84.3	$\geqslant 1.1 \times 10$[illegible]	$\geqslant 3.3 \times 10$[illegible]	$1.3 \cdot 10$[illegible]	3.0×10[illegible]	6.6×10[illegible]
^{176}Hf → ^{172}Yb	$0^+ \to 0^+$, g.s.	$\geqslant 9.3 \times 10^{19}$	–	2.5×10^{20}	6.6×10^{20}	2.0×10^{20}
	$0^+ \to 2^+$, 78.7	$\geqslant 1.8 \times 10^{16}$	$\geqslant 3.0 \times 10^{17}$	1.3×10^{22}	3.5×10^{22}	4.9×10^{21}
^{177}Hf → ^{173}Yb	$7/2^- \to 5/2^-$, g.s.	$\geqslant 3.2 \times 10^{20}$	–	4.5×10^{20}	5.2×10^{22}	4.4×10^{22}
	$7/2^- \to 7/2^-$, 78.6	$\geqslant 7.5 \times 10^{16}$	$\geqslant 1.3 \times 10^{18}$	9.1×10^{21}	1.2×10^{24}	3.6×10^{23}
^{178}Hf → ^{174}Yb	$0^+ \to 0^+$, g.s.	$\geqslant 5.8 \times 10^{19}$	–	3.4×10^{23}	1.1×10^{24}	2.2×10^{23}
	$0^+ \to 2^+$, 76.5	$\geqslant 6.9 \times 10^{16}$	$\geqslant 2.0 \times 10^{17}$	2.4×10^{25}	8.1×10^{25}	7.1×10^{24}
^{179}Hf → ^{175}Yb	$9/2^+ \to 7/2^+$, g.s.	$\geqslant 2.5 \times 10^{20}$	$\geqslant 2.2 \times 10^{18}$	4.5×10^{29}	4.0×10^{32}	4.7×10^{31}
	$9/2^+ \to 9/2^+$, 104.5	$\geqslant 5.5 \times 10^{17}$	$\geqslant 2.2 \times 10^{18}$	2.0×10^{32}	2.5×10^{35}	2.2×10^{34}
^{180}Hf → ^{176}Yb	$9/2^+ \to 7/2^+$, g.s.	–	–	6.4×10^{45}	5.7×10^{46}	9.2×10^{44}
	$9/2^+ \to 9/2^+$, 82.1	–	$\geqslant 1.0 \times 10^{18}$	4.0×10^{49}	4.1×10^{50}	2.1×10^{48}

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Conclusion

- To study the decay of naturally occurring hafnium to the ground state and the first excited state of a CHC crystal scintillator was used in coincidence with a HP-Ge detector in 2848 h of live time.

- The results rules out the $T_{1/2}$ value of the decay of ^{174}Hf given in literature. In particular, we found that the decay of ^{174}Hf to the ground state has been definitely observed with a **$T_{1/2}$ = 7.0(1.2) x 10^{16} y**. This value is in good agreement with the theoretical predictions.

- lower limits of the half-life for decay of ^{174}Hf to the first excited state and for decay of ^{176}Hf, ^{177}Hf, ^{178}Hf, ^{179}Hf either to the ground state or to the first excited level of daughter nuclides (10^{16}-10^{20} y).

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